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13. ABSTRACT (Maximum 200 words)

The technology of second harmonic generation in channel waveguides made from poled polymers was developed for interaction geometries appropriate for (1) cascading and wavelength shifting, and (2) for wavelength demultiplexing. For fundamental beams which generate a harmonic along the same propagation direction, various phase-matching geometries were studied which relied on periodically modulating the linear refractive index and/or the nonlinearity (QPM) in DANS. Although this approach with interdigitated poling electrodes did produce the best results ever reported in polymeric systems, it was not competitive with those in the ferroelectric QPM-LiNbO$_3$. The best results were obtained using modal phase-matching in which a low order fundamental waveguide mode was velocity matched to a higher order harmonic mode. By using multiple layers with successive layers poled in opposite directions, a figure of merit equal to LiNbO$_3$ was achieved in the polymer DR1. However, the large losses at the second harmonic reduced the effective sample length and hence the absolute conversion efficiency. For oppositely propagating fundamental inputs, a geometry appropriate to demultiplexing, alternatively poled layers led to the largest conversion efficiencies for any material system for second harmonic beams radiated normal to the waveguide surfaces.

14. SUBJECT TERMS polymer science, second harmonic generation in polymer waveguides, nonlinear polymers, polymer devices, polymer waveguides, polymer multi-stack poling

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Title of Project: Novel Frequency Conversion Phenomena Based on Poled Polymers

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Subject Terms: Nonlinear optics in polymers, nonlinear polymers, nonlinear organic devices, second harmonic generation, poled polymers

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Research Progress:

This program consisted of two separate components.

1. <u>Waveguide Second Harmonic Generation with Poled Polymers: Towards Cascading and Parametric Devices</u>

The ultimate program goal was to demonstrate cascading devices at communications wavelengths with poled polymers. The immediate goal was to obtain efficient second harmonic generation (SHG) in channel waveguides at 1.5μm. We used two well-known side-chain polymers with the active chromophore consisting of either DANS or DR1. Alignment of the second-order active species was achieved by parallel plate poling at nominal fields of 50-100 V/μm, much less than the optimum values of >300 V/μm we had demonstrated previously. Underlying this work was the challenge to show that poled polymers could be competitive with the benchmark doubling material, periodically poled $LiNbO_3$. Our initial target was the Figure of Merit (FOM) in terms of %/W-cm². It is defined in terms of $\eta = P(2\omega)/P^2(\omega)\text{-}L^2$ which quantifies the conversion for a 1 cm long (L=1), phase-matched channel waveguide. A second figure of merit, ultimately the most important one, was $\eta' = P(2\omega)/P^2(\omega)$, the absolute conversion efficiency for the sample. Frequently the absorption limits the effective length of the sample.

Efficient SHG requires phase-matching which means that momentum (wavevector) must conserved in the nonlinear interaction between the participating waves. The biggest challenge faced was to find a technique which utilizes the strengths of poled polymer technology. Towards this end, we assessed essentially two techniques for wavevector matching. We started with Quasi-Phase-Matching (QPM) which was the key to the success of $LiNbO_3$. When the limitations of this approach became clear for the polymer case, we investigated modal dispersion phase matching (MDPM) which made optimum use of the poling techniques.

A detailed analysis was performed of the SHG figures of merit for polymers as a function of wavelength of the fundamental. Evaluated were both the FOM and the absolute conversion efficiency. The polymer DANS was used as the example, mainly because we have detailed

information about the wavelength variation of the nonlinearity as well as the losses. (Unfortunately such complete data is available for very few polymers!) In brief summary, we found that DANS is only useful for incident wavelengths between about 1250 and 1650 nm due to the large losses, primarily at the doubled wavelength. Finally, we found that the two-level model was completely inadequate in evaluating the FOM that could be expected from a given polymer. The failure was primarily in using the two-level model for describing the wavelength dependence of the absorption coefficient.

The experimental achievements can be summarized in terms of the following table which tracks our progress for the FOM over the duration of the program. The details will be discussed later. The key point is that a FOM equivalent to $LiNbO_3$ has indeed been demonstrated.

Mode Conversion	# layers	L_{pm} mm	$\alpha(\omega)$ dB/cm	$\alpha(2\omega)$ dB/cm	FOM $W^{-1}cm^2$
$TM_{00}(\omega) \rightarrow TM_{00}(2\omega)$ QPM (1995) DANS	3	0.6	40		0.01%
$TM_{00}(\omega) \rightarrow TM_{00}(2\omega)$ QPM (1995) DANS	3	2	5	30	.05%
$TM_{00}(\omega) \rightarrow TM_{20}(2\omega)$ MPM (1996) DANS	3	2	5	50	1%
$TM_{00}(\omega) \rightarrow TM_{10}(2\omega)$ MPM (1996) DANS*	4	>7	6.5	20	14%
$TM_{00}(\omega) \rightarrow TM_{20}(2\omega)$ MPM (1997) DR1**	5 invert	>7	3.5	95	44%
$TM_{00}(\omega) \rightarrow TM_{00}(2\omega)$ QPM (1997) $LiNbO_3$	NA	24	0.4	>0.6	43%

* ½ DANS, ½ polyetherimide in guiding film

** alternate layers have different T_gs

Three different quasi-phase-matching geometries were investigated. The idea in QPM is to modulate either the refractive index or the nonlinearity $d^{(2)}$ with a periodicity Λ so that $k_2 - 2k_1 \pm 2\pi/\Lambda = 0$ where k_2 and k_1 are the propagation wavevectors for the harmonic and fundamental respectively . Λ is adjusted so that this condition is satisfied at a given wavelength. In polymers, we investigated three waves of fabricating such a

periodicity.

(i)   Photobleaching with blue light reduces locally the refractive index and the second order nonlinearity in the side-chain polymer DANS so that photobleaching through a periodic mask introduces a modulated index grating. A $d^{(2)}$-active film, sandwiched between two buffer layers, was first fabricated by parallel plate poling, the top electrode was removed, a single mode channel waveguide and a periodic index and nonlinearity modulation were photobleached into the film. Maker fringe measurements established a 25 pm/V nonlinearity for a nominal poling voltage of only 100V/μm in the unbleached regions. The linear propagation losses at the fundamental were measured to be very large, 25 dB/cm. Both by modeling and direct observation we found that the losses are due to coherent coupling to radiation fields via the modulated index structure, i.e. for the fundamental the index modulation acts like a phased array antenna coupling the guided light into radiation fields. Under these difficult conditions, the conversion efficiency for a 2mm long waveguide was 5×10^{-4}%/W. Nevertheless, the losses were just too high for useful SHG.

(ii)   Periodic modulation of the nonlinearity was achieved by fabricating the DANS-buffer multistack film between two electrodes, one planar and the other spatially periodic (fingers). In the first case, the periodic electrodes were deposited on the upper surface of the buffer film. During poling, the compressive forces due to the strong electric fields cause the finger electrodes to sink into the polymer film, leading to large material distortions. Depressions of order of 5% of the total film thickness have been measured. This in turn also leads to large fundamental propagation losses, of order 40 dB/cm.

(iii)   The material distortion was reduced by more than one order of magnitude by depositing the finger electrodes on the substrate, and the flat continuous electrode on the upper film surface. In this case the losses were only 4 dB/cm (including coupling losses) for 2.5 mm long samples. In this case we were able to achieve phase-matching over the entire channel length. We measured a figure of merit of 0.05%/W-cm² for this geometry. Because the effective modulated nonlinearity in our poling geometry was only a few pm/V, we abandoned this approach. The problem was the overall stack thickness of 4-5 μm which did not allow an efficient modulation due to spatial spreading of the poling field.

Implementing modal phase matching (MPM) was the key to the large improvement in the SHG FOM. This approach relies on using waveguides which are single mode at the fundamental (1550 nm) and multimode at the SHG wavelength (always the case in waveguide SHG). Thus modal phase-matching (which requires that the phase velocity of the fundamental and harmonic fields be equal) can in principle be implemented with one of a number of second harmonic waveguide modes. Given the wavelength dispersion in the refractive index due to both the materials and the confinement properties of the waveguide, the fundamental can be the lowest order mode but the SH *must* be a higher order spatial mode. The MPM technique was applied to a sequence of single and multilayer films with different properties designed to sequentially improve the SHG performance.

(i)   With a nominal poling field of 100 V/μm (parallel plate poling), MPM was implemented in a single unidirectionally-poled DANS whose thickness was chosen for the $TM_{00}(\omega)$ to $TM_{20}(2\omega)$ mode phase-matching combination. This led to an

improvement of the FOM to 1%/W-cm^2 and a phase-matching length of about 2 mm was inferred. This an improvement by a factor of 20 over our best previous results obtained by QPM (quasi-phase-matching).

In modal phase-matching, the efficiency is proportional to the product of the nonlinearity d$^{(2)}$, the fundamental field squared and the harmonic field, integrated over the waveguide cross-section.

$$FOM = \frac{P(2\omega,L_{eff})}{P^2(\omega,0)L_{eff}^2} \propto \int\int dxdy d^{(2)}(x,y)E^2(\omega,x,y)E(2\omega,x,y)$$

where E(ω) and E(2ω) are the fundamental and harmonic fields respectively. For higher order second harmonic modes, the sign of the field changes one or more times across the waveguide core region. Noting that the fundamental field squared is always positive, interference occurs in this integral reducing the conversion efficiency for a uniform d$^{(2)}$. This was the case for the TM$_{00}$(ω) to TM$_{20}$(2ω) combination just discussed. This interference can be reduced by using d$^{(2)}$-inactive layers in the region from which negative contributions would originate, or it can be eliminated by reversing the sign of the nonlinearity in those regions.

(ii)   MPM was implemented for the TM$_{00}$(ω) to TM$_{10}$(2ω) mode combination by making approximately one half of the guiding layer from DANS, and the second half from a SH-inactive material, in this case polyetherimide. The TM$_{10}$(2ω) field reverses its sign once across the guiding film so that contributions from a uniform guiding core will tend to cancel. However, because the polyetherimide is "dead" for SHG, no cancellation occurs which eliminates the negative contribution. The resulting FOM was 15%/W-cm^2. This at the time was the best value reported in channel polymeric waveguides to date, considering the λ^{-4} dependence of the conversion efficiency on the fundamental wavelength. (It was superceded by our own later work to be discussed next.) This value was also competitive for the first time with the best reported LiNbO$_3$ waveguide values of 22%/W-cm^2 at that time in 1996. However, the absolute conversion efficiency [$\propto \alpha^{-2}(2\omega)$] is not yet competitive because of the large losses at the harmonic wavelength, as discussed previously.

A fortuitous ratio between the loss coefficients at the fundamental and harmonic wavelengths allowed us to estimate limitations on the phase-matching length. When this ratio is approximately two, the SHG spectral bandwidth is directly proportional to the phase-matching length, independent of losses. We found our coherence length to be 7 mm, the same as the sample length. This implies that the phase-matching length, usually limited by waveguide fabrication dimensional tolerances, was larger than 7 mm.

(iii)   We have established a collaboration with Siegfried Bauer (of the University of Potsdam and the Heinrich Hertz Institute) to investigate the reverse poling option for MPM. This approach involves reversing the sign of the nonlinearity (and therefore the net poling) whenever the harmonic field reverses sign so that the product d$^{(2)}$(x) E(2ω,x) is always positive and hence contributions from all of the waveguide sections add constructively. This was implemented with a two step poling process using two DR1-

based polymers with different glass transition temperatures separated by about 27^0C. The polymer films consisted of spun on layers with alternating layers of high T_g and low T_g material. The full stack was first poled near the high T_g temperature, and then repoled with the applied field reversed at the lower T_g. As a result only the low T_g film was (re-)oriented in the reversed field because the orientation of the chromophores in the high T_g polymer was locked in at such reduced temperatures. For "n" films, the nonlinearity was reversed "n-1" times. In particular, prototype samples with two oppositely poled regions were made for $TM_{00}(\omega)$ to $TM_{10}(2\omega)$ SHG, and three layers with alternating molecular orientations for $TM_{00}(\omega)$ to $TM_{20}(2\omega)$ phase-matching. A variety of non-invasive characterization techniques were used to verify the reverse poling in the successive films. This produced the best results to date, as summarized in the table.

With advances in our polymer film processing technology, namely:

1. production of very uniform films;
2. multilayer coating and poling of materials with different glass transition temperatures T_g to specific tolerances; and
3. the successful development of the modal phase matching technique with successive, oppositely poled films,

our best result with the DANS polymer is currently 15%/W-cm^2 with a phase-matching length of 7 mms and 44%/W-cm^2 in multilayers of DR1 polymer with again a phase-matching length of about 7 mm. These FOM results are comparable to the best value in LiNbO$_3$ channel waveguides of 44%/W-cm^2 at 1500 nm and phase-matching over 2.5 cms, published in early 1997. Our latest MDPM waveguides were poled with (i) a nominal 5 V/μm across each active layer, and (ii) cladding layers with conductivities not optimized relative to the nonlinear material. Hence the 5 pm/V operative in the latest experiments should be extendible to > 200 pm/V with a better choice of materials. It is noteworthy that the LiNbO$_3$ values are almost at their theoretical limit, where-as the poled polymers still have at least one order of magnitude improvement possible. However, the absolute device conversion efficiency, which is proportional to $\alpha(2\omega)^{-2}$ where $\alpha(2\omega)$ is the second harmonic loss coefficient in cm^{-1}, is limited to a few percent primarily by "killer" losses at the second harmonic frequency of 10s-100 dB/cm in the polymers used. The principal barriers to better performance are:

1. the large propagation losses in DANS (~15 dB/cm) and DR1 (>100 dB/cm) encountered at 750-800 nm;
2. to a much lesser degree the propagation losses at 1550 nm of 3-4 dB/cm in both polymer systems.

Research has been started on alternate materials including some polyurethanes from SUNY Buffalo, based on chromophores such as 4-[-N-(2-hydroxyethyl)-N—(methyl)aminophenyl]-4'-(6-hydroxyhexyl sulfonyl) stilbene, hereafter called APSS. The absorption peak of this dye is located at a shorter wavelength compared to azo compounds or stilbene derivatives with a nitro group as the electron withdrawing group. As a consequence this peak is strongly blue shifted compared to the DANS we are currently using. Losses of the order of 1 dB/cm have been measured in unpoled waveguides at 823 nm. Other characteristics being comparable to DANS, very high net efficiency should be achieved with such a material. If this chromophore proves to be successful, different polymeric environments will be studied. In particular a first version

of a crosslinkable polymer has already been developed and could be taken advantage from. Further improvements could be made to the material.

We have analyzed the improvement in η and η' possible with improved materials and higher net poling fields. One option that we are currently pursuing is in-plane poling. In principle, the full voltage occurs across the active material, in a direction parallel to the surfaces.

Figure: $d^{(2)}_{eff}$ in pm/V necessary to obtain, at the optimal length, a given net SHG efficiency (% /W) as a function of second harmonic waveguide losses. The device
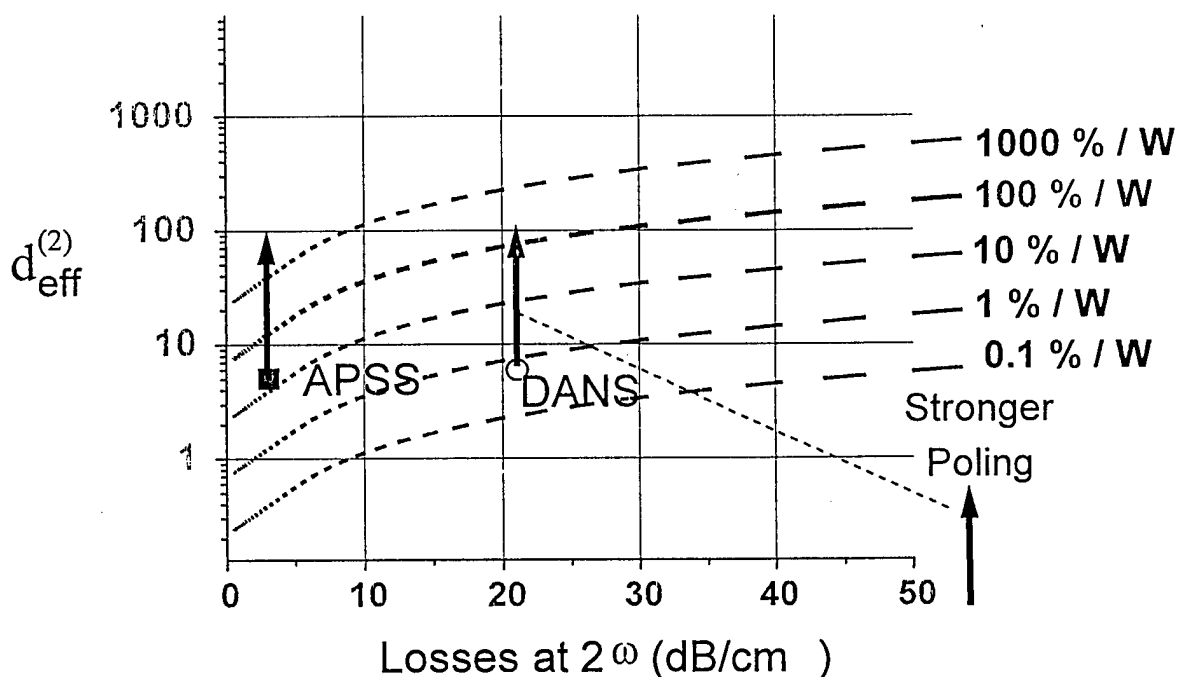
structure assumed is an active-inactive layer combination. The actual experimental device is referred by a o, a similar device using APSS as active material is identified by ■. The vertical arrows give an indication of the area that could be reached by optimizing the poling conditions.

Thus FOMs ⇨ 2000%/W-cm² with materials comparable to DANS are possible.

2. <u>Demultiplexing Based on Counter-Propagating SHG With Poled Polymers</u>

The ultimate goal was to build a wavelength demultiplexer based on sum and difference frequency generation by oppositely propagating beams. The immediate goal was to optimize the interaction cross-section for sum frequency generation by oppositely propagating input beams in a poled polymer channel waveguide. This interaction geometry leads to a sum frequency signal generated at an angle to the surface normal, the angle being proportional to the difference between the input frequencies.

On the theoretical side, a detailed analysis was performed of the figures of merit for counter-propagating, fundamental, guided waves for polymers as a function of wavelength of the fundamental. Evaluated were both the FOM and the absolute conversion efficiency. Again, the polymer DANS was used as the example. For this geometry we found that the signal levels were optimized for materials in which the sum frequency wavelength was approximately equal to the λ_{max} of the polymer.

The major achievement was a demonstrated enhancement of the cross-sections for this process by using multilayer stacks for quasi-phase-matching in the direction normal to the waveguide surface (the direction into which the signal is radiated). This was achieved by spinning on successive layers of buffer (L) and DANS (NL) layers, producing a stack in which every second layer is second-order active. The layer thicknesses were chosen to be one half wavelength at the sum frequency. When poled with in-plane electrodes, the nonlinearity was modulated from 0 to $d^{(2)}_{max}$.

Experiments on improving the efficiency by transverse QPM were performed with NL/L multilayer devices consisting of 3-layer DANS-SCP films as the nonlinear layers with layers of PC-polymer films in between them as the linear layers. The samples poled at 200 V/μm and 300 V/μm were investigated and their nonlinear cross sections were determined to be 1.2×10^{-6} W^{-1} and 2.3×10^{-6} W-1 respectively. The largest nonlinear cross section to date yields the power conversion efficiency for this geometry of $\eta = P(2\omega)/LP_+(\omega)P_-(\omega) = 0.6\%$/W-cm. This means that for a 1 watt input, 6 mW are obtained in a 1 cm sample (our sample length).

These values are only 20% of the theoretically calculated numbers. Although these large discrepancies are not fully explained at this time, some possible reasons will be given here. There can be increased propagation losses due to the multilayer structure. Strong stray scattering was observed in the tapered region, especially at the end of the taper. The higher order mode is close to cut-off for the multilayer waveguide. It is possible that a part of the beam is coupled into the higher order mode and scattered out by the narrow channel waveguide. Another possible reason for the discrepancy is an inhomogeneity of the nonlinearity across the poling electrode gap. Although the inhomogeneity and the influence of the charge injection were determined as minimal for the film poled at 50 V/μm, a larger influence of charge injection can occur for films poled at higher fields. The nonlinearity in the middle of the gap where the channel waveguide is located can be smaller than the value evaluated in the measurement because the measured values are averaged over the 20 μm electrode gap. An over-estimate of the nonlinearity leads to an over-estimated nonlinear cross section in the theoretical calculation. Nevertheless, despite the discrepancy between the measurement and the theory, there is a large improvement in the conversion efficiency. The enhanced SH light was easily detected by an uncooled CCD.

These experiments established that a wavelength demultiplexer based on this interaction is feasible and the signal levels are reasonable. With improvements in both processing and materials, we estimate that 6 mW outputs with 100 mW inputs should be possible.

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        #"Lightwave manipulation in guided wave geometries: $\chi^{(2)}$", ICONO'2, Japan, July 1995
        #"Second Harmonic Generation with Poled Polymers", MRS Meeting, Boston, November 1995
        #"Sum Frequency Generation in Composite Polymers", China-USA Workshop on Composite Materials, Nanjing China, April 1996
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        #"Progress Towards WDM Demultiplexing with Sum and Difference Frequency Generation in Poled Polymers", Fourth International Conference on Frontiers in Polymers and Advanced Materials, Cairo Egypt, January 1997
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        #"Poling and Characterization of Photonic Waveguide Devices for Efficient Second-Harmonic Generation", (given by Matthias Jaeger), SPIE Symposium, San

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#"Second Harmonic Generation in Multilayer Poled Polymer Waveguides", Annual OSA Meeting, October 1997

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